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NEWS

02

Polymer Composites

Versatile materials find new uses in many fields

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Cover: Technicians at Sikorsky manufacturing facility oversee production of helicopter rotor blade made of hybrid composite of Kevlar and graphite fibers. Photo courtesy of Du Pont

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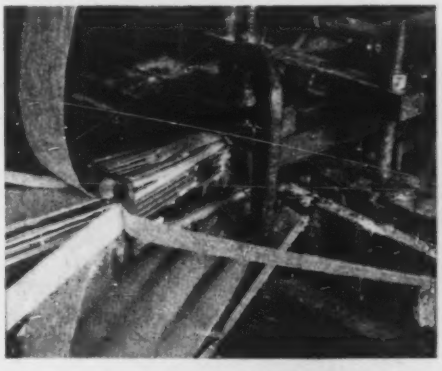


News Focus

The growth of advanced polymer composites is spurred by a wide variety of applications in products that were formerly made of more traditional materials, such as metals. **Page 7**

Wide Variety of Applications Spark Polymer Composites Growth

Advanced composites are one aspect of an evolving polymer composites universe that includes a heterogeneous mixture of materials, forms, and processes

Ward Worthy, C&EN Chicago

Question: What do the following items have in common: bowling lane, pressure cooker, canoe, and motor home?

Answer: Not much—except that they're all among the items that won product excellence awards at the Composites Show in Cincinnati last month. Needless to say, the four items were all made entirely or largely of "composites." Moreover, there were 36 other award winners in 11 different categories—plus many, many nonwinners.

The point is that composites—specifically, polymeric composites—are being used nowadays to make hundreds of types of products that formerly were the domain of more traditional materials, notably metals. The list includes airplanes, boats, auto parts, chemical process equipment, appliance cabinets, business machine housings, even caskets.

In many cases, composites can't compete with metals on a cost-per-pound basis. However, composite parts are often lighter than their metal counterparts, so fewer pounds are required. Also, the composites are inherently corrosion resistant and thus may last longer. Moreover, they present opportunities for savings through "parts consolidation." Often, a large complex part can be molded in one piece, whereas to make the corresponding part of metal would require the fabrication of many separate components that then would have to be assembled—an expensive process.

As is often the case with words, the word composite means different things to different people. In its broadest sense, a composite is simply a material containing more than one component. But to "materials people," the word has a narrower sense. They restrict its use to materials in which some type of reinforcement is embedded in some type of matrix. They also speak of synergy: The composite, desirably, has properties superior to the properties of its components taken separately.

Boat weighing 18% less than a typical boat of its size is made of nonwoven continuous glass fiber reinforcements

There are several kinds of composites, some with metal matrices, some with ceramic matrices, and some with matrices made of an organic polymer. With polymer-based composites (the subject of this article), the reinforcing fibers and fillers add tensile strength and stiffness. They often also improve dimensional stability—that is, they help reduce shrinkage during parts fabrication.

The polymer matrix does a lot more than hold the reinforcements together. It also plays the major role in determining such important matters as resistance to chemicals and weathering, thermal and electrical behavior, and appearance. Often it also adds toughness and impact resistance.

In addition to composites, there are "advanced composites." Although that term has lately attained buzzphrase status, its meaning is a little fuzzy, too.

Materials engineers define advanced composites quantitatively: modulus so much, moment of inertia so much, elongation-to-failure so much. Undoubtedly, their definitions are accurate, but they don't mean much to the uninitiated.

Price might be a good way to define them. Advanced composite systems based on thermoset resins currently cost about $30 to $60 a lb. Some of the newer advanced thermoplastic systems now under development cost $100 to $200 a lb. Amazingly, for some applications, they're worth it.

Perhaps the best definition: Advanced composites are composite materials that, by virtue of their outstanding strength-to-weight properties, make it possible to do things that otherwise couldn't be done.

For instance, Bryan Allen wouldn't have been able to pedal Paul MacCready's Gossamer Albatross across the English Channel in 1979. Similarly, Dick Rutan and Jeana Yeager wouldn't have been able to fly their Voyager nonstop around the world last December. Both aircraft, each a pioneer in its own way, owed much of their success to the strong, lightweight advanced composites that made up most of their airframes.

Looking ahead, a project that excites the advanced composites community is the Air Force's advanced tactical fighter (ATF) program. Two teams—one consisting of Lockheed, General Dynamics, and Boeing, the other of Northrop and McDonnell—are competing to build prototypes. According to John Wanamaker, a materials and process group engineer with Lockheed, probably about 50% of the ATF's structural weight will be composite materials, compared to less than 10% in today's fighter planes.

Not only are composites thus evolving technologically, they also make up a growth industry. The latter conclusion is borne out by figures compiled recently by the Composites Institute, an arm of the Society of the Plastics Industry.

Overall, shipments of composites in 1986 came to a record 2.28 billion lb, a 2.8% increase from 1985's 2.2 billion lb. For 1987, shipments are projected to grow another 3.0%, to nearly 2.35 billion lb (C&EN, Feb. 9, page 5).

Carl Rue, general chairman of the institute, notes that most segments of the composites industry have been growing faster than the economy as a whole. And that above-average growth rate, he says, appears likely to go on.

Rue, who is also vice president of sales and marketing for CertainTeed Corp. (a manufacturer of glass fiber for reinforcements), points to the auto industry—the largest single market for composites—as an example. Although U.S. auto production in 1986 fell 3.7% from the 1985 level, composites shipments to the industry rose 3.0% from 1985 levels, to 585 million lb. This year, auto production is expected to fall again. Even so, composites consumption is expected to increase another 4.0%, to 608 million lb. Furthermore, Rue says, the average composite content per vehicle will grow from 50 lb today to 61 lb by 1991.

The aircraft/aerospace/military market for composites is small, in terms of pounds, but it grew a whopping 12% in 1986, with shipments rising to 37 million lb. Another strong year is forecast for 1987, with shipments increasing 8.1% to 40 million lb. And they tend to be high-priced pounds, thanks to a large proportion of advanced composites.

Among the large-poundage markets, the construction industry is the second largest user (after autos). Shipments to that market totaled 456 million lb in 1986, a 2.7% increase from 1985. This year, Rue says, housing starts likely will drop. Nevertheless, he sees a 1.8% increase to 464 million lb.

In the marine market, composites shipments totaled 340 million lb, up 3.0% from 1985. In 1987, shipments are expected to grow another 3.2%, to 351 million lb.

After a flat 1985, composites use in the electric/electronics market rebounded in 1986. Shipments were up 5.8%, to 201 million lb. Growth will continue this year, with shipments expected to rise another 3.5% to 208 million lb.

Longer term, the outlook for composites is also rosy. According to a recent market analysis made by Kenneth E. Jacobson, a senior consultant with consulting firm Charles H. Kline & Co., sales of composites to certain specialty markets will grow from $2.5 billion in 1986 to more than $3.7 billion in 1991. That translates to an average annual increase of 8%, compounded.

The Kline study divides the market into different categories from those used by SPI's Composites Institute in its report of shipments. The former includes filled as well as reinforced composites; the latter includes primarily reinforced composites. Conversely, Jacobson points out, the study omits such major markets as construction and corrosion-resistant equipment, whereas the Composites Institute figures include those markets. Nevertheless, the conclusions reached by the two reports are generally in agreement.

For example, "automotive applications" accounted for about $850 million, or 34% of the total, of composite sales in 1986. The market will grow 7% a year, reaching $1.2 billion annually by 1991, the Kline study says. Electronics applications amounted to about $750 million, or 30% of the total, in 1986. That market is expected to grow 9% a year, to just under $1.2 billion annually by 1991.

The "other transportation" category, which includes aerospace, marine, and truck applications, will be the fastest-growing market of all. According to the Kline study, that market, which amounted to about $425 million in 1986, will grow an average 13% annually over the next five years, reaching about $780 million by 1991.

Anthony J. Cardinal, Du Pont vice president for textile fibers, is looking even farther ahead: "We see composites as a major growth market, with an annual value of about $10 billion by the late 1990s." He adds that Du Pont intends to concentrate its own efforts on advanced composites, where, he says, consumption has been increasing at an annual rate of 30% for the past decade.

To describe the world of composites and some of the shifts taking place in its evolution is to describe a

universe of heterogeneous complexity. A multitude of components—resins, reinforcements, and additives—go into composites. The composite raw materials can take a wide variety of forms. And many differing processes are used to fabricate the materials into parts.

A list of all the resins that can be used to make composites is, for all practical purposes, a list of all resins. Not surprisingly, however, some resins are used more than others.

For example, in the Kline study of composites in specialty markets, total consumption of filled and reinforced plastics came to 2.4 billion lb in 1986. Of that, thermoset resin-based composites accounted for about 1.5 billion lb (62.5%) and thermoplastic resin-based composites for 900 million lb (37.5%).

Among the thermosets, polyesters were first, with about 60% of the total, followed by phenolics with 16% and epoxies with 15% of the total. "Others" accounted for the remaining 9%. Of the thermoplastics, polypropylene ranked first, with 44% of the total, followed by nylons with 26% and thermoplastic polyesters with 14%. "Others" accounted for the remaining 16%.

It's no accident that the thermoset polyesters have such a commanding lead. They offer a good balance of properties, are amenable to most fabrication processes, and are relatively inexpensive. Phenolics are more heat resistant and less flammable than polyesters, and they're also inexpensive. However, they're not so good cosmetically.

Epoxies, by virtue of their chemical and thermal resistance, strength, and dimensional stability, are usually the resins of choice for the so-called advanced composites. However, they're relatively slow-curing, and that characteristic limits their use in high-volume production applications. Polyurethanes, polyureas, polyisocyanurates, and related thermoset polymers are widely used in reinforced reaction injection molding (RRIM) and resin transfer molding (RTM). Among the newer thermoset resins, vinyl esters are related to both epoxies and polyesters. They're said to provide the toughness and corrosion resistance of the former and the processing ease of the latter.

"Hybrid resins" are now being made, using a two-step process. First, a hydroxyl-terminated unsaturated isophthalate polyester is synthesized. That polymer is then reacted with a diisocyanate and styrene. The resulting hybrids are said to be tougher than polyesters and stronger, stiffer, and less expensive than polyurethanes.

A somewhat related approach involves the creation of an "interpenetrating polymer network" (IPN), in which two or more polymers are synthesized in the immediate presence of one another. The goal, again, is a product whose properties are superior to those of its individual components. For example, an acrylic-isocyanate IPN is said to provide exceptional resistance to moisture and high temperatures.

At the high-performance end of the spectrum are polyimides and bis-maleimides. They are among the few thermoset resins that provide the high strength, stiffness, and particularly the heat resistance required for advanced composites applications.

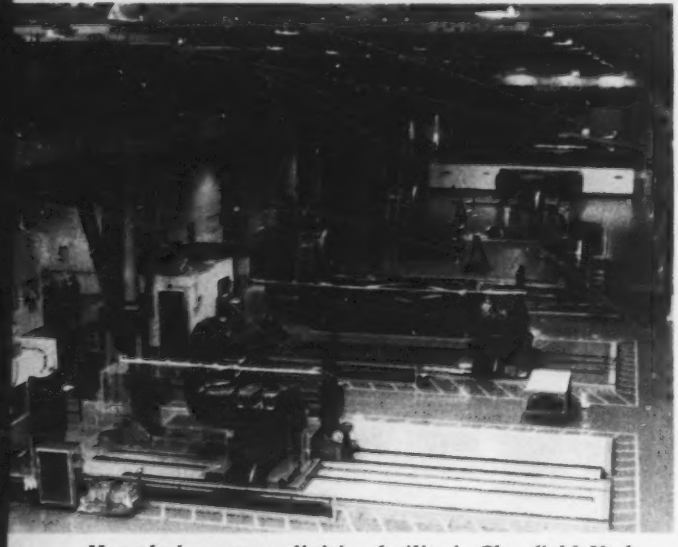
Although the thermosets have a substantial head start in the composites race, the thermoplastics are moving up fast. Their performance is in many cases as good as that of thermosets, and they offer some advantages in terms of handling and fabrication. For one thing, cycle times can be shorter, a decided advantage in high-volume applications. Also, if a part made of a thermoset composite is somehow botched during fabrication, it's usually just scrap. In contrast, a part made of a thermoplastic composite often can be remelted and reformed. That's especially desirable with the high-priced resins and fibers used for advanced composites.

Polypropylenes, the volume leader among the thermoplastics, probably would get more respect if they cost more. As it is, they offer, at a relatively low price, a good balance of properties, light weight, and exceptional resistance to flexing and impact.

Nylons and thermoplastic polyesters can compete with many of the thermosets in terms of electrical, dimensional, and thermal characteristics. They also tend to be more impact-resistant than the thermosets, and they're easier to fabricate. Acetals, fluoropolymers, polycarbonates, polyphenylene oxide, styrenic copolymers, and vinyls are among the other resins that have found one or more niches in the world of engineering thermoplastics.

In addition, several relatively new thermoplastic resins with outstanding mechanical and thermal properties are competing for the high end of the advanced composites market. The candidates include polyamide-imides, polyetherimides, polyetherether ketones, polyphenylene and polyarylene sulfides, and polyether sulfones.

Just as any of many resins may be used to form the matrix of a composite, so may any of a host of materi-



Hercules' aerospace division facility in Clearfield, Utah, applies filament winding for advanced composites to make missile engine cases

als be used to fill or reinforce the matrix. A corollary: Just as a composite may contain more than one type of resin, so may it contain more than one type of filler or reinforcement.

Composites may be of several major types, generally described as filled, laminar ("sandwich"), fiber-reinforced, or as combinations thereof. In fact, the categories don't break down that nicely, there being a sort of continuum. Generally speaking, the greater the aspect (length to diameter) ratio of the filler or fiber, the greater the reinforcing effect, in terms of increased tensile strength and stiffness. That is, flakes provide more reinforcement than spherical particles. Short fibers yield stronger products than flakes. Composites reinforced with long fibers are stronger than those reinforced with short fibers. "Advanced" composites typically are reinforced with continuous lengths of high-strength fibers.

Variety of applications of polymeric composites includes Voyager aircraft, auto front end, satellite tracking antenna, and bowling lane surface

In the case of fiber-reinforced composites, the arrangement of the fibers also affects the strength. If all the fibers point in one direction, the composite is strongest in that direction. If fibers are arranged with some at an angle to the others (as in a woven fabric), the strength is apportioned in both directions. If the fiber arrangement is random or multidirectional, the part tends to be about equally strong in all directions. Obviously, the choice of arrangements would be determined by the properties desired. "Put the fibers where the loads are" is the general rule.

There are two other important rules for fiber composites, according to industry consultant Gerald D. Shook. One, the modulus of the fiber should be greater than the modulus of the matrix. Two, the elongation of the fiber should be less than the elongation of the matrix. Shook uses this analogy to explain the rules: "You don't reinforce concrete with rubber hose."

Hundreds of different materials are used as fillers for composites. Talc, calcium carbonate, kaolin, wood flour, and mica are a few of the more widely used. One major reason for using fillers is to reduce costs. Generally, the fillers are a lot cheaper than the resins they extend. The fillers may even be air or other gases, as in the case of structural foams.

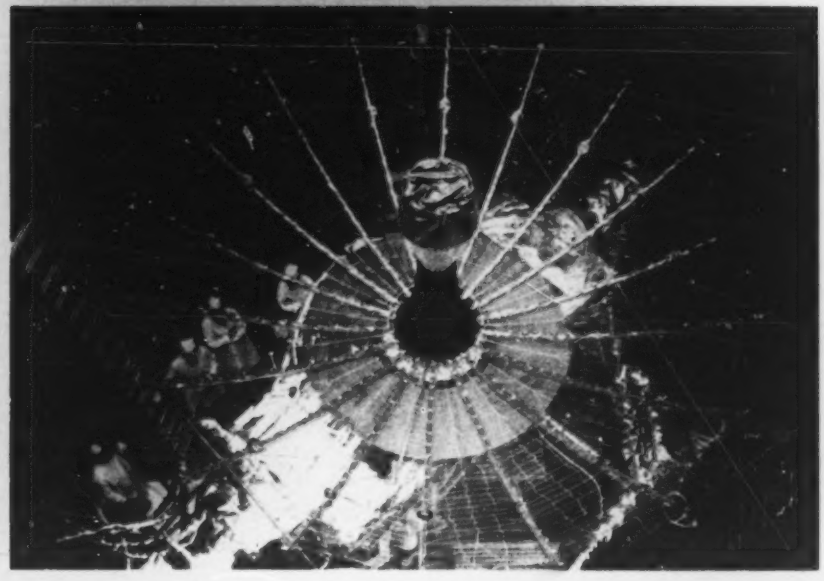
That doesn't mean that the fillers degrade the product, however. They can also add many useful properties. For example, they can increase its compression strength (just as sand and gravel increase the compression strength of concrete). They can improve the thermal and electrical properties of the composite. Or they can increase its dimensional stability. In the case of composites filled with flaky materials (like mica), with aspect ratios of the order of 25- or 50-to-1, the fillers also make a modest contribution to tensile strength and stiffness as well.

In common parlance, the term "reinforced plastics" usually refers to products consisting of resin matrices stiffened and strengthened by the addition of fibers. Such everyday items as sisal and cotton fibers and fabrics, and even paper, find application as "crack stoppers."

However, the real workhorse among the reinforcing fibers is undeniably the borosilicate glass fiber known as "E-glass." It provides an excellent balance of properties at reasonable cost. It's been available commercially for more than half a century. And a great deal of technology has formed around it. A number of makers provide it in a variety of lengths and forms: chopped or milled short fibers, rovings (bundles of parallel strands—sort of untwisted yarn or rope), mats, woven or knitted fabrics, and others.

Although E-glass is the predominant reinforcing fiber, it isn't the only one. For example, polymers can form not only the matrix of a composite but also the reinforcing fibers. Special high-strength grades of polyester and nylon 6 fibers are offered for that purpose, for use either alone or together with glass fibers. Compared with glass, the thermoplastic fibers are lighter, less brittle, and tougher. According to Allied-Signal, it is currently the only U.S. firm supplying these fibers in forms specifically engineered for composite reinforcement.

In addition, several "high-performance" fibers have



been developed in recent years (C&EN, Feb. 2, page 9). These fibers have properties that set them apart from the rest. In particular, they have outstanding strength-to-weight characteristics and/or high resistance to heat and chemical attack.

Although the high-performance fibers weren't necessarily developed to reinforce composites, a number of them are being used for that purpose, either experimentally or commercially. Currently, these fibers are quite expensive, so that their use is largely restricted to the advanced composites.

Carbon (graphite) fibers, Owens Corning's S2-glass fibers, and Du Pont's Kevlar aramid fibers are the fibers now most widely used for advanced composites. Spectra fiber, an extended-chain polyethylene fiber developed by Allied-Signal, is a relative newcomer to the composites scene. Generally, the carbon and glass fibers have the edge in terms of stiffness and heat resistance, whereas the polymer fibers excel in terms of light weight and toughness. In some applications, both types may be used together to provide an optimum balance of properties.

Resins and reinforcements may be the two main components of composites. But there can be and generally is more to a composite than just those two. Among other constituents that, depending on needs, may or may not be included in a given composite: accelerators, binders, flame retardants, flexibilizers, inhibitors, low-profile agents, low-shrink agents, mold release agents, pigments, sizings, suppressants, thickeners, thixotropes, ultraviolet absorbers, and wetting agents.

In other words, composites can be complicated. But many of the people who make things out of composites don't want to be in the formulation business. They want to buy a product that they can put in their machines and fabricate and that will do what it is supposed to do. Consequently, the putting together and selling of composite "systems" is a business unto itself.

Just as there are many resins and reinforcements available, so are there many different composite "compounds." In many instances, they're designed to use with specific fabrication processes, or to meet specific product requirements. For example, the resin-to-fiber ratio can be varied over a fairly wide range.

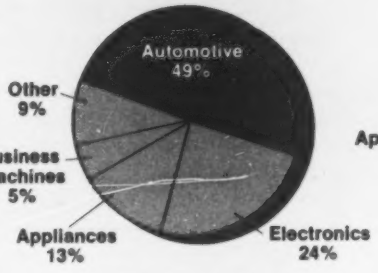
One type of compound is the "prepreg," which is short for "product made by preimpregnating woven or continuous reinforcement fibers with resins." Prepregs can be made with either thermoset or thermoplastic resins. And the prepreg may also include one or several additives.

Thermoset prepregs are brought to a relatively dry condition by partial curing. The final cure takes place during fabrication, with the application of additional heat. Carbon/epoxy and aramid/epoxy prepregs are often used for advanced composite applications. Some thermoset prepregs need to be refrigerated during shipment and storage, to prevent premature curing. Thermoplastic prepregs don't.

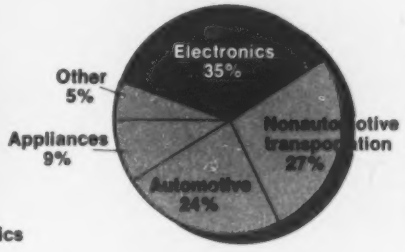
Sheet molding compound (SMC) is used for a variety of flat, thin parts (like auto body panels and appliance cabinets). To make it, chopped fibers, thermoset resin, fillers, and additives are spread on a film of polyethylene. A second polyethylene film is placed on top. This "sandwich" is kneaded to mix the components, pressed into a sheet about $\frac{1}{4}$ inch thick, and then wound into a roll.

There are variations on the SMC theme. Thick molding compound, for example, is similar to SMC, but thicker (up to 2 inches). It's too thick to wind into rolls, so it's usually cut into slabs of appropriate size. Another variation, unidirectional SMC, incorporates continuous as well as chopped roving and thus provides greater directional strength and stiffness. Thermoplastic sheets can be made by a melt lamination process, using either chopped strand or continuous strand mats.

Auto and electronics industries are biggest markets for composites . . .

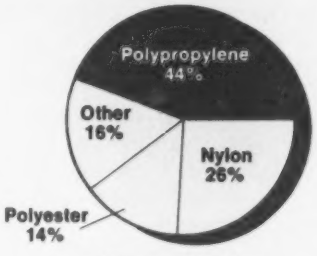


Thermoplastics
Total = $1.1 billion

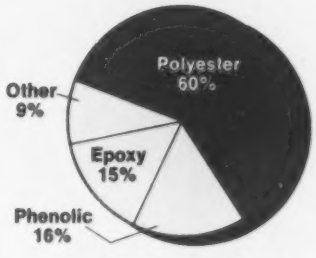


Thermosets
Total = $1.4 billion

. . . and polypropylene and polyester are most widely used resins



Thermoplastics
Total = 900 million lb



Thermosets
Total = 1.5 billion lb

Note: Consumption of filled and reinforced plastics in specialty markets, 1986. **Source:** Charles H. Kline & Co.

Bulk molding compound (BMC) combines short fibers, resins, and additives into a doughy material. It may be furnished in bulk, as the name suggests, or it may be extruded into "logs" for easier handling. One purveyor notes that BMC is used "where fine finish, good dimensional stability, part complexity, and good overall mechanical properties are important."

Many short-fiber compounds are furnished in pellet form for injection molding applications. These may be based on either thermoset or thermoplastic resins. And, like the other types of compound, they may contain a variety of additives, including lubricants to ease their passage into the mold.

As the variety of composite systems suggests, there are many processes that may be used to turn the materials into finished parts.

Perhaps the simplest of the processes is hand layup. Reinforcing fibers are arranged in a mold, a thermoset resin is poured over the fibers, and the mixture is pressed into shape with rollers. Then the setup is left alone while the resin cures, usually at room temperature.

The sprayup process is similar to hand layup, except that a special type of spray gun chops the fiber and mixes it with the resin, then sprays the mixture into the mold. The sprayup approach allows more complexly shaped parts to be molded than does hand layup.

With both of these open mold processes, it's common practice, for the sake of appearance, to apply a resin-rich, pigmented "gel coat" to the mold before applying the main resin-fiber mixture. Both processes are best suited for low- to medium-volume production of large parts, including such things as boats, tanks, and building panels. Both processes provide only one finished surface. Equipment costs are relatively low, but skilled operators are essential to success.

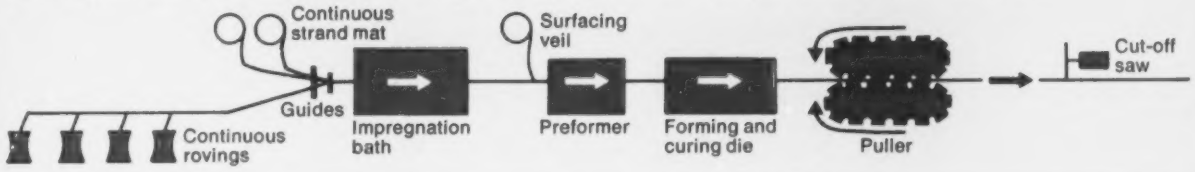
For slightly higher production rates, and for situations where two finished surfaces are required, a number of low-pressure closed molding processes are available. The simplest of these is cold press molding, which is a lot like the open molding processes except that the mold has a top as well as a bottom.

With resin transfer molding (RTM), the reinforcement is placed in the bottom half of a two-part mold and the mold is closed. Then the resin is pumped in. Reinforced reaction injection molding (RRIM) is similar, except that separate streams of chemicals are pumped into the mold, where the polymerization reaction then takes place. To make "structural" RRIM parts, long-fiber reinforcements are arranged in the mold before the liquids are pumped in, much as with RTM. For less demanding applications, flakes or milled fibers can be mixed with one of the liquid streams before it's pumped into the mold.

Compression molding processes are usually called for where high-volume production is involved. Or to put it another way, equipment costs are so much higher for these processes that high volumes are necessary to justify them. However, labor costs are substantially lower. The methods are suitable for thermoset composites (often using sheet or bulk molding compounds) and also for thermoplastic composites. In contrast to the open-mold and low-pressure closed-mold processes, external heat is added to hasten curing of thermosets, or to soften or melt thermoplastics so they will flow into the mold.

Injection molding processes can produce large numbers of complex parts reinforced with fillers or short fibers. The techniques can be used with either thermoset or thermoplastic resin systems, but with modifications. For thermoplastics, the injection chamber must be hot and the mold cool. For thermosets,

Pultrusion enables continuous molding of simple or complex structural shapes

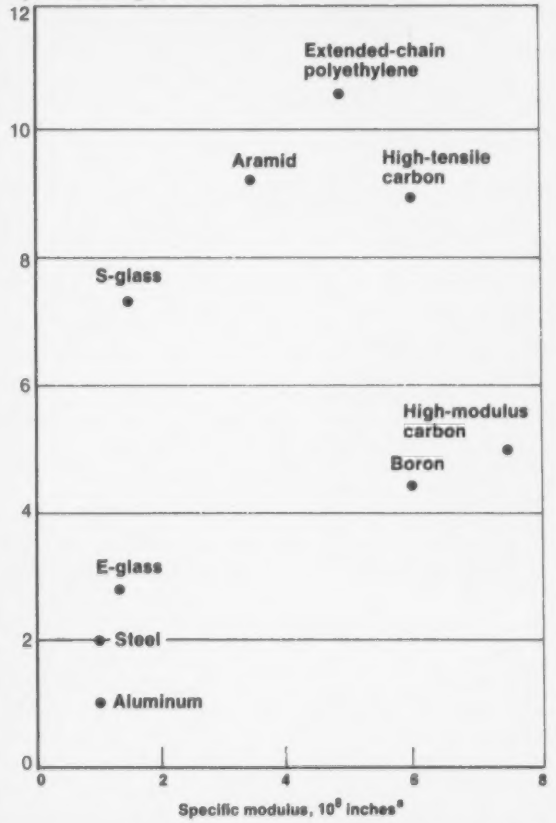


however, the injection chamber must be cool and the mold hot.

High-volume fabrication processes such as injection molding are suitable only for use with short fibers. Conversely, only the relatively slow techniques requiring manual arrangement of the fibers are suitable for products reinforced with long continuous fibers.

Yet how advanced the composites are depends in large part on the fact that they employ continuous fibers oriented to afford maximum strength and stiffness. Indeed, a shortage of suitable fabrication techniques has hindered the broader use of advanced composites, especially in cost-conscious mass production industries like the auto industry.

High-performance reinforcing fibers have good strength-to-weight properties



Note: Values are representative. [a] Strength or modulus divided by density.

According to J. Michael Bowman, director of Du Pont's composites division, fabrication is the "Achilles' heel" of advanced composites. Even for the most demanding aerospace and military applications, he points out, the advanced composite materials often must be formed into parts with essentially the same hand layup techniques that are used to make boats.

The process is further complicated by the fact that the resins used in these systems cure at high temperatures, so that the parts must be autoclaved. Consequently, development of fast, automated production techniques for advanced composites has a high priority.

However, two fairly well-automated processes now in use are specifically designed for continuous fibers. One of these processes is filament winding. Continuous lengths of strand or roving are impregnated with resin, then wound onto a rotating mandrel in a pattern calculated to provide maximum strength where it's needed. The process is used to make hollow, more or less cylindrical parts like chemical storage tanks, pipes, rocket motor cases, helicopter rotor blades, and automotive drive shafts.

The other long-fiber process is known as pultrusion. As the name suggests, pultrusion is a combination of extrusion and pulling. Rovings, continuous strand mat, tapes, or other appropriate reinforcements are pulled through tanks of thermoset resins, through a preformer, and then through a die, where the product is formed into its final shape and cured by heating. The process is used for the continuous manufacture of a variety of structural shapes—including complex ones—of constant cross section: beams, pipes, bars, rails, and such.

Finally, just as development of fabrication processes for composites has lagged behind the development of materials, so has the science of composites lagged behind the technology of composites. Much more is known than is understood.

Some progress is being made, however, in industry as well as in academia. For example, organizations devoted to basic research in composites have been established at several universities. Probably the best known is the University of Delaware Center for Composite Materials, founded in 1974. Similar centers have been established at other schools, including Virginia Polytechnic Institute, Purdue, and Rensselaer Polytechnic Institute. Generally, funds to support these activities are furnished by the composites industry, by the National Science Foundation, and by other government agencies with an interest in composites. □